

10/568344

IAP20 Rec'd PCT/PTO 14 FEB 2006

**THE FOLLOWING ARE THE ENGLISH TRANSLATION  
OF ANNEXES TO THE INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT (ARTICLE 34):**

Amended Sheets (Pages 59-70)

10/568344

AS ENCLOSED TO IPRP

IAP20 Rec'd C/IPTO 14 FEB 2006

**We claim**

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1. The use of uncharged transition metal complexes of the formula (I) comprising at least one carbene ligand in organic light-emitting diodes



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where the symbols have the following meanings:

M<sup>1</sup> is a metal atom selected from the group consisting of Co, Rh, Ir, Nb, Pd, Pt, Fe, Ru, Os, Cr, Mo, W, Mn, Tc, Re, Cu, Ag and Au in any oxidation state possible for the respective metal atom;

carbene is a carbene ligand which may be uncharged or monoanionic and monodentate, bidentate or tridentate, with the carbene ligand also being able to be a biscarbene or triscarbene ligand;

20

L is a monoanionic or dianionic ligand, which may be monodentate or bidentate;

25

K is an uncharged monodentate or bidentate ligand selected from the group consisting of phosphines; phosphonates and derivatives thereof, arsenates and derivatives thereof; phosphites; CO; pyridines; nitriles and conjugated dienes which form a π complex with M<sup>1</sup>;

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n is the number of carbene ligands, where n is at least 1 and when n > 1 the carbene ligands in the complex of the formula I can be identical or different;

35

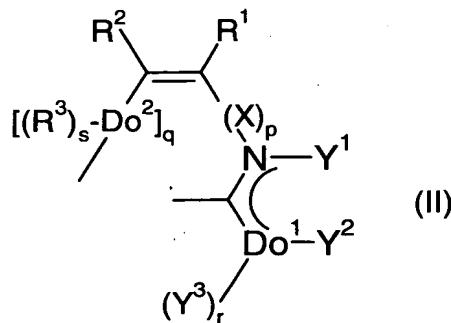
m is the number of ligands L, where m can be 0 or ≥ 1 and when m > 1 the ligands L can be identical or different;

o is the number of ligands K, where o can be 0 or ≥ 1 and when o > 1 the ligands K can be identical or different;

where the sum  $n + m + o$  is dependent on the oxidation state and coordination number of the metal atom and on the denticity of the ligands carbene, L and K and also on the charge on the ligands carbene and L, with the proviso that n is at least 1.

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2. The use as claimed in claim 1, wherein the uncharged transition metal complexes are employed as emitter molecules.
  
3. The use as claimed in claim 1 or 2, wherein the carbene ligand or ligands is/are bidentate.
  
4. The use as claimed in any of claims 1 to 3, wherein the carbene ligand or ligands is/are monoanionic.
  
- 15 5. The use as claimed in any of claims 1 to 4, wherein the carbene ligand or ligands has/have the formula II



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where the symbols have the following meanings:

$Do^1$  is a donor atom selected from the group consisting of C, P, N, O and S preferably, P, N, O and S;

25  $Do^2$  is a donor atom selected from the group consisting of C, N, P, O and S;

$r$  is 2 when  $Do^1$  is C, is 1 when  $Do^1$  is N or P and is 0 when  $Do^1$  is O or S;

$s$  is 2 when  $Do^2$  is C, is 1 when  $Do^2$  is N or P and is 0 when  $Do^2$  is O or S;

30

$X$  is a spacer selected from the group consisting of silylene, alkylene, arylene, heteroarylene and alkenylene;

$p$  is 0 or 1;

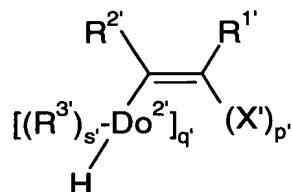
q is 0 or 1;

5  $Y^1, Y^2$  are each, independently of one another, hydrogen or a carbon-containing group selected from the group consisting of alkyl, aryl, heteroaryl and alkenyl groups;

or

10  $Y^1$  and  $Y^2$  together form a bridge between the donor atom  $Do^1$  and the nitrogen atom N which has at least two atoms of which at least one is a carbon atom,

$Y^3$  is hydrogen or an alkyl, aryl, heteroaryl or alkenyl radical;  
or



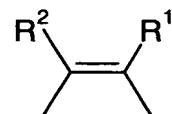
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where  $Do^2$ ,  $q'$ ,  $s'$ ,  $R^3'$ ,  $R^1'$ ,  $R^2'$ ,  $X'$  and  $p'$  are each, independently of one another, as defined for  $Do^2$ ,  $q$ ,  $s$ ,  $R^3$ ,  $R^1$ ,  $R^2$ ,  $X$  and  $p$ ;

20  $R^1, R^2$  are each, independently of one another, hydrogen or an alkyl, aryl, heteroaryl or alkenyl radical,

or

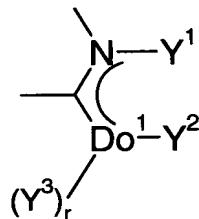
25  $R^1$  and  $R^2$  together form a bridge having a total of from three to five atoms of which one or two atoms may be heteroatoms and the remaining atoms are carbon atoms, so that the group



30 forms a five- to seven-membered, ring which may contain, in addition to the existing double bond, one further double bond or in the case of a six- or seven-membered ring two further double bonds and may be substituted by alkyl or aryl groups and may contain at least one heteroatom;

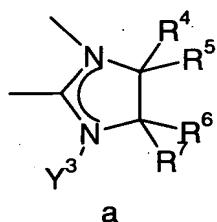
35  $R^3$  is hydrogen or an alkyl, aryl, heteroaryl or alkenyl radical.

6. The use as claimed in claim 5, wherein the group

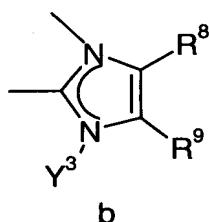


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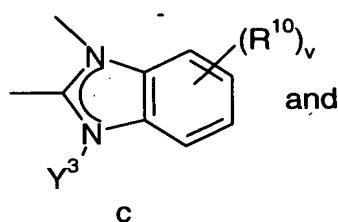
is selected from the group consisting of



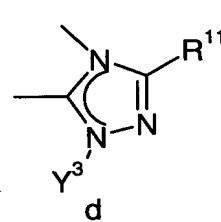
a



b



c



d

10

where the symbols have the following meanings:

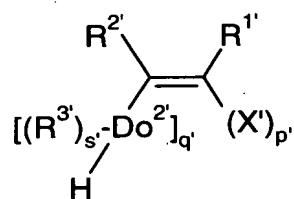
$R^4, R^5, R^6,$   
 $R^7, R^8, R^9$

15 and  $R^{11}$  are each hydrogen, alkyl, aryl, heteroaryl or alkenyl or a substituent which acts as a donor or acceptor;

$R^{10}$  is alkyl, aryl, heteroaryl or alkenyl or 2 radicals  $R^{10}$  together form a fused-on ring, or  $R^{10}$  is a radical which acts as a donor or acceptor;

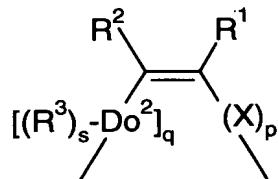
20  $v$  is from 0 to 4 and when  $v$  is 0 the four carbon atoms of the aryl radical in the formula c which may be substituted by  $R^{10}$  bear hydrogen atoms;

$Y^3$  is a hydrogen atom or an alkyl, aryl, heteroaryl or alkenyl radical;  
 25 or



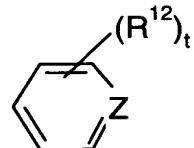
where  $D_0^{2'}$ , q, s,  $R^3'$ ,  $R^1'$ ,  $R^2'$ , X and p are defined, independently of one another, as for  $D_0^2$ , q, s,  $R^3$ ,  $R^1$ ,  $R^2$ , X and p.

- 5 7. The use as claimed in claim 5 or 6, wherein the group



denotes the structure

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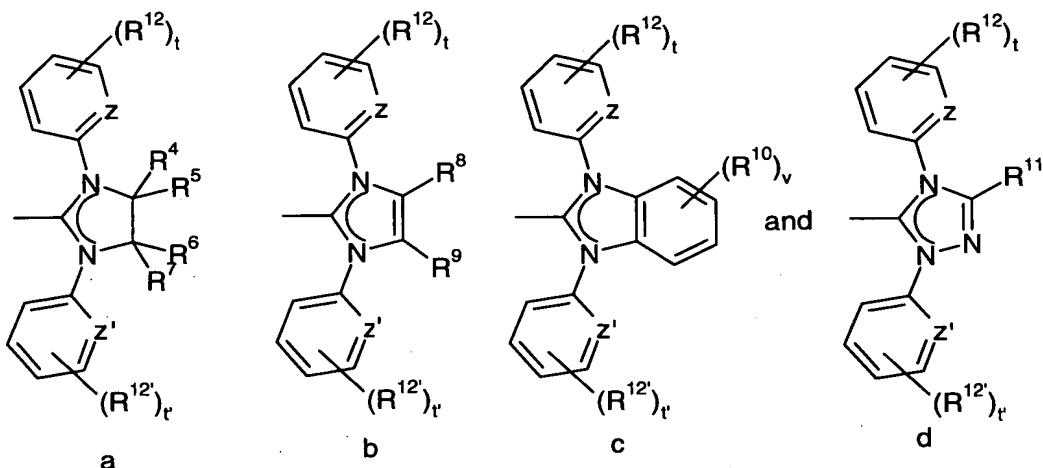
where the symbols have the following meanings:

15 Z is CH or N and can be located in the o, m or p position relative to the point of linkage of the group to the carbene ligand;

20  $R^{12}$  is an alkyl, aryl, heteroaryl or alkenyl radical, or 2 radicals  $R^{12}$  together form a fused-on ring which may contain one or more heteroatoms, or  $R^{12}$  is a radical which acts as a donor or acceptor;

t is 0 to 3 and when t > 1 the radicals  $R^{12}$  can be identical or different.

25 8. The use as claimed in any of claims 5 to 7, wherein the carbene ligand or ligands is/are selected from the group consisting of



where the symbols have the following meanings:

- 5        Z, Z'      are identical or different and are each CH or N;

10      R<sup>12</sup>, R<sup>12'</sup> are identical or different and are each an alkyl, aryl, heteroaryl or alkenyl radical, or 2 radicals R<sup>12</sup> or R<sup>12'</sup> together form a fused-on ring which may contain at least one heteroatom, or R<sup>12</sup> or R<sup>12'</sup> is a radical which acts as a donor or acceptor;

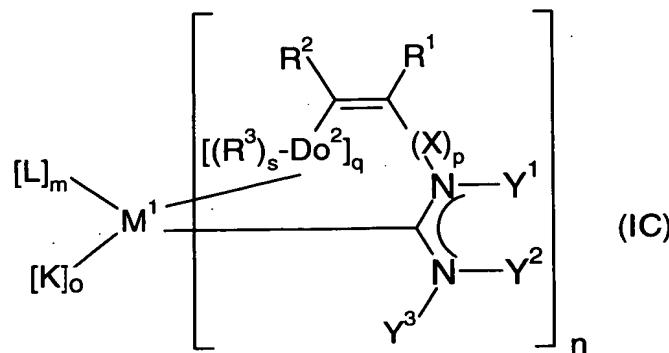
15      t and t' are identical or different and are each from 0 to 3, and when t or t' > 1 the radicals R<sup>12</sup> or R<sup>12'</sup> can be identical or different;

20      R<sup>4</sup>, R<sup>5</sup>, R<sup>6</sup>,  
          R<sup>7</sup>, R<sup>8</sup>, R<sup>9</sup>  
          and R<sup>11</sup> are each hydrogen, alkyl, aryl, heteroaryl or alkenyl or a radical which acts as a donor or acceptor;

25      R<sup>10</sup>     is alkyl, aryl, heteroaryl or alkenyl or 2 radicals R<sup>10</sup> together form a fused-on ring which may contain at least one heteroatom, or R<sup>10</sup> is a radical which acts as a donor or acceptor;

v        is from 0 to 4 and when v is 0 the four carbon atoms of the aryl radical in the formula c which may be substituted by R<sup>10</sup> bear hydrogen atoms.

9. An uncharged transition metal complex of the formula  $\text{IC}$



where the symbols have the following meanings:

5       $M^1$       is Ru, Rh, Ir, Pt in any oxidation state possible for the respective metal atom;

L      is a monoanionic or dianionic ligand, which may be monodentate or bidentate

10     K      is an uncharged monodentate or bidentate ligand;

n      is the number of carbene ligands, where n is at least 2 and the carbene ligands in the transition metal complex can be identical or different;

15     m      is the number of ligands L, where m can be 0 or  $\geq 1$  and when  $m > 1$  the ligands L can be identical or different;

20     o      is the number of ligands K, where o can be 0 or  $\geq 1$  and in the case of  $o > 1$  the ligands K can be identical or different;

where the sum  $n + m + o$  is dependent on the oxidation state and coordination number of the metal atom used and the denticity of the ligands and also on the charge on the ligands, with the proviso that n is at least 2;

25      $Dº²$       is a donor atom selected from the group consisting of C, N, P, O and S;

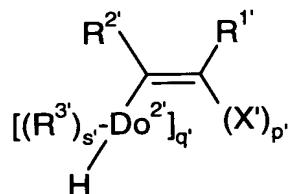
s      is 2 when  $Dº²$  is C, is 1 when  $Dº²$  is N or P and is 0 when  $Dº²$  is O or S;

30     X      is a spacer selected from the group consisting of silylene, alkylene, arylene, heteroarylene and alkenylene;

p      is 0 or 1;

q is 0 or 1;

Y<sup>3</sup> is hydrogen or an alkyl, aryl, heteroaryl or alkenyl radical;  
5 or

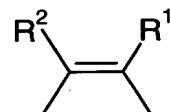


10 where Do<sup>2</sup>, q', s', R<sup>3</sup>, R<sup>1</sup>, R<sup>2</sup>, X' and p' are each, independently of one another, as defined for Do<sup>2</sup>, q, s, R<sup>3</sup>, R<sup>1</sup>, R<sup>2</sup>, X and p;

R<sup>1</sup>, R<sup>2</sup> are each, independently of one another, hydrogen or an alkyl, aryl, heteroaryl or alkenyl radical,

or

15 R<sup>1</sup> and R<sup>2</sup> together form a bridge having a total of from three to five atoms of which one or two atoms may be heteroatoms and the remaining atoms are carbon atoms, so that the group

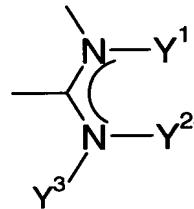


20 forms a five- to seven-membered ring which may contain, in addition to the existing double bond, one further double bond or in the case of a six- or seven-membered ring two further double bonds and may be substituted by alkyl or aryl groups and may contain heteroatoms, or the ring is fused to further rings which may contain one or more heteroatoms;

25 R<sup>3</sup> is hydrogen or an alkyl, aryl, heteroaryl or alkenyl radical;

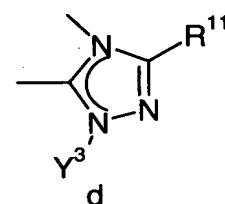
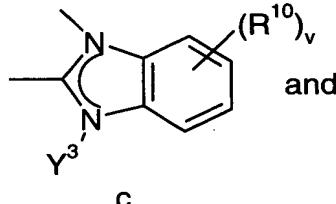
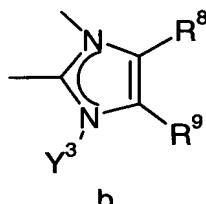
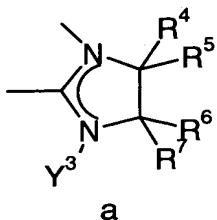
30 Y<sup>1</sup>, Y<sup>2</sup> together form a bridge between the nitrogen atoms N which has at least two atoms of which at least one is a carbon atom, where the bridge can be saturated or unsaturated and the two or more atoms of the bridge may be substituted or unsubstituted and when the bridge has two carbon atoms and is saturated at least one of the two carbon atoms is substituted.

10. A transition metal complex as claimed in claim 9, wherein the group



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is selected from the group consisting of



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where the symbols have the following meanings:

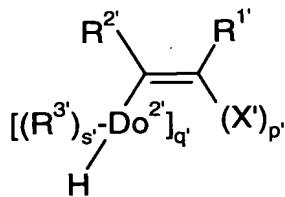
$R^4$ ,  $R^5$ ,  $R^6$ ,  
 $R^7$ ,  $R^8$ ,  $R^9$

15 and  $R^{11}$  are each, independently of one another, hydrogen, alkyl, aryl, heteroaryl or alkenyl, where at least one of the radicals  $R^4$ ,  $R^5$ ,  $R^6$  and  $R^7$  in the formula a is not hydrogen, or a radical which acts as a donor or acceptor;

20  $R^{10}$  is alkyl, aryl, heteroaryl or alkenyl or 2 radicals  $R^{10}$  together form a fused-on ring which may contain one or more heteroatoms, or  $R^{10}$  is a radical which acts as a donor or acceptor;

v is from 0 to 4 and when v is 0 the four carbon atoms of the aryl radical in the formula c which may be substituted by  $R^{10}$  bear hydrogen atoms;

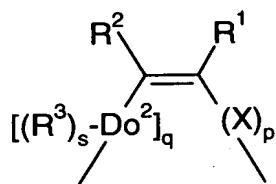
25  $Y^3$  is a hydrogen atom or an alkyl, aryl, heteroaryl or alkenyl radical; or



where  $\text{Do}^2$ ,  $q'$ ,  $s'$ ,  $R^3'$ ,  $R^1'$ ,  $R^2'$ ,  $X'$  and  $p'$  are each, independently of one another, defined as for  $\text{Do}^2$ ,  $q$ ,  $s$ ,  $R^3$ ,  $R^1$ ,  $R^2$ ,  $X$  and  $p$ .

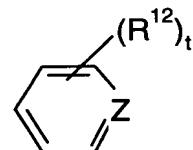
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11. A transition metal complex as claimed in claim 9 or 10, wherein the group



10

denotes the structure



where the symbols have the following meanings:

15

$Z$  is CH or N and can be located in the o, m or p position relative to the point of linkage of the group to the carbene ligand;

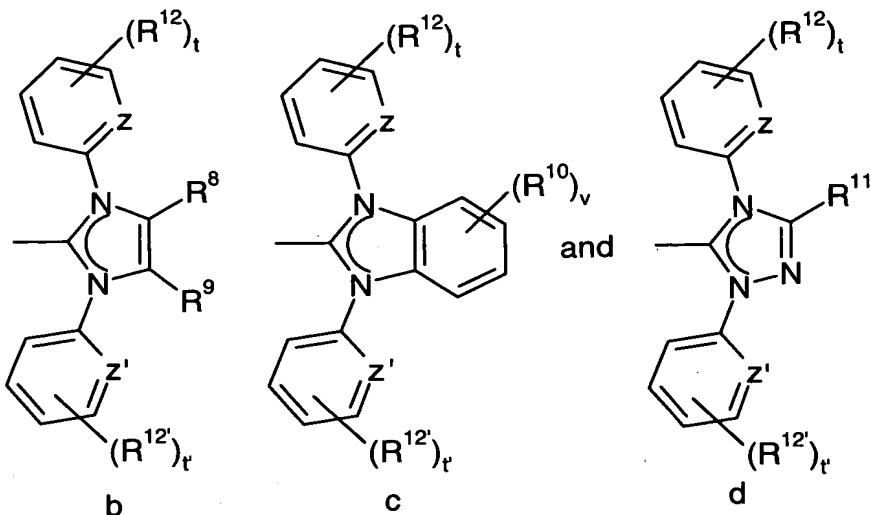
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$R^{12}$  is an alkyl, aryl, heteroaryl or alkenyl radical, or 2 radicals  $R^{12}$  together form a fused-on ring which may contain one or more heteroatoms, or  $R^{12}$  is a radical which acts as a donor or acceptor;

$t$  is 0 to 3 and when  $t > 1$  the radicals  $R^{12}$  can be identical or different.

25

12. A transition metal complex as claimed in any of claims 9 to 11, wherein the two or more carbene ligands are selected independently from the group consisting of



where the symbols have the following meanings:

- 5       $Z, Z'$     are identical or different and are each CH or N;
- 10      $R^{12}, R^{12'}$  are identical or different and are each an alkyl, aryl, heteroaryl or alkenyl radical, or 2 radicals  $R^{12}$  or  $R^{12'}$  together form a fused-on ring which may contain at least one heteroatom, or  $R^{12}$  or  $R^{12'}$  is a radical which acts as a donor or acceptor;
- 15      $t$  and  $t'$  are identical or different and are each from 0 to 3, and when  $t$  or  $t' > 1$  the radicals  $R^{12}$  or  $R^{12'}$  can be identical or different;
- 20      $R^8, R^9$   
and  $R^{11}$  are each hydrogen, alkyl, aryl, heteroaryl or alkenyl or a radical which acts as a donor or acceptor;
- 25      $R^{10}$     is alkyl, aryl, heteroaryl or alkenyl or 2 radicals  $R^{10}$  together form a fused-on ring which may contain at least one heteroatom, or  $R^{10}$  is a radical which acts as a donor or acceptor;
- v       is from 0 to 4 and when v is 0 the four carbon atoms of the aryl radical in the formula c which may be substituted by  $R^{10}$  bear hydrogen atoms.
13. A transition metal complex as claimed in any of claims 9 to 12, wherein  $M^1$  is Ir(III),  $n$  is 3 and  $m$  and  $o$  are each 0, with the three carbene ligands preferably being identical.

14. A process for preparing transition metal complexes as claimed in any of claims 9 to 13 by the deprotonation of the ligand precursors corresponding to the appropriate carbene ligands and subsequent reaction with suitable metal complexes in which the desired metal is present.

5

15. An OLED comprising at least one transition metal complex as defined in any of claims 1 to 8 or as claimed in any of claims 9 to 13.

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16. A light-emitting layer comprising at least one transition metal complex as defined in any of claims 1 to 8 or as claimed in any of claims 9 to 13.

17. An OLED comprising a light-emitting layer as claimed in claim 14.

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18. A device selected from the group consisting of stationary VDUs such as VDUs of computers, televisions, VDUs in printers, kitchen appliances and advertising signs, lighting units, information signs, and mobile VDUs such as VDUs in mobile telephones, laptops, vehicles and destination displays on buses and trains comprising an OLED as defined in claim 15 or 17.